ABSOLUTE FREQUENCY MEASUREMENTS OF THE 28- AND 78- μ m cw WATER VAPOR LASER LINES

K. M. Evenson, J. S. Wells, L. M. Matarrese, and L. B. Elwell Radio Standards Physics Division, National Bureau of Standards, Boulder, Colorado 80302 (Received 10 November 1969; in final form 5 January 1970)

The two highest frequency measurements as yet reported are described. Frequencies of the 28- and $78-\mu m$ cw water vapor laser lines were found to be 10.718073 ± 0.000002 THz and 3.821775 ± 0.000003 THz, respectively, by beating each of these radiations with the 337- and 373- μm (0.89- and 0.80-THz) radiation from an HCN laser, in a metal-on-metal point-contact diode. The frequencies of the HCN laser were in turn measured by beating the 337- μm radiation with 74-GHz radiation and by measuring the 337- and 373- μm frequency difference.

This letter describes the two highest frequency measurements as yet reported. The measurements of these laser frequencies advance the progress toward frequency measurements in a region of the electromagnetic spectrum where precise wavelength measurements are being made. 1,2 The simultaneous measurement of both frequency and wavelength may provide a defined value for the speed of light. Recent progress in extending frequency measurement techniques toward the visible region of the spectrum include absolute frequency measurements of the $118-\mu\mathrm{m}^3$ and $84-\mu\mathrm{m}^4$ $\mathrm{H_2O}$ and $\mathrm{D_2O}$ laser lines.

The frequencies of the 28- and 78- μ m (10.7- and 3.8-THz) cw water vapor laser lines were

measured by beating each of these lines with 337-and 373- μ m (0.89- and 0.80-THz) radiation from an HCN laser. The harmonic generation and mixing occurred in a tungsten-on-nickel catwhisker diode. The over-all technique resembled that used by Hocker et al. When they measured the frequencies of the 118- and 84- μ m H₂O and D₂O lines. The chief difference was the substitution of the metal-on-metal diode for the metal-on-silicon diode.

Figure 1 indicates the over-all experimental arrangement. The two 8-m-long lasers used compound beam-splitter coupling. 7 The HCN laser had a confocal mirror geometry and a 10-cm diam, and the $\rm H_2O$ laser had a folded confocal geometry

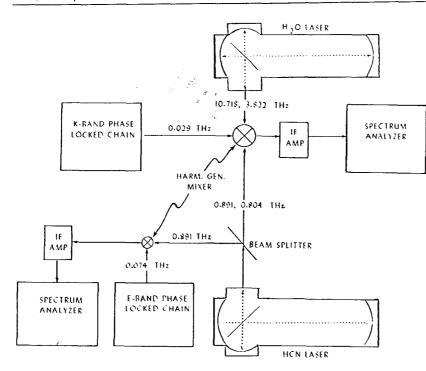


FIG. 1. Block diagram of 28-and $78-\mu m$ frequency measuring experiment.

and 4.1-cm diam. The HCN laser ran on $\mathrm{NH_3}$ and $\mathrm{CH_4}$ at a pressure which produced striations about 2.5 cm apart in the discharge. With a discharge current of 600 mA, the laser produced a power output of 50 mW at 0.89 THz. The water vapor laser was filled with $\mathrm{H_2O}$ and $\mathrm{H_2}$ at approximately 0.9 Torr. With the current at 250 mA it produced about 75 mW at 10.7 THz, and considerably less at 3.8 THz. The 29- and 74-GHz radiations were made up from the output of phase-locked klystron chains. These frequencies were measured by an x-band frequency counter, which was compared with NBS standard frequencies.

An open structure was utilized to support the tungsten-on-nickel harmonic generator-mixer point-contact diode. A 1-mil-diam tungsten catwhisker 2.5 mm long was etched to a diameter of about one-half mil for two-thirds of its length, sharpened conventionally, and placed in contact with the polished end face of a piece of nickel welding rod, 1.6 mm in diameter. The pressure of the contact was adjustable by means of the differential screw that held the rod. Radiation from the three sources impinged on the catwhisker in the three directions appropriate for maximum coupling of each, in agreement with antenna theory. 8

The catwhisker of the diode was connected to a wide-band i.f. amplifier passing 1 to 60 MHz with a 4-dB noise figure, and a gain of 58 dB. The amplifier was in turn connected to a spectrum analyzer for visual observation of the beat notes. The initial observation of the beat note from the 10.7-THz water vapor line was made with the

spectrum analyzer replaced by a 120-kHz bandwidth communications receiver with its output connected to a phase-sensitive detector referenced to a 600-Hz chopper on the water vapor laser.

A one-half mil polyethylene beam splitter deflected a few percent of the HCN beam into a conventional cross-guide harmonic generator so that the 0.89-THz frequency could be continuously monitored by observing the beat between the 12th harmonic of a phase-locked 74-GHz klystron and the HCN laser line.

Although the experiment was designed to beat the 28- μ m line with the 12th harmonic of the $337-\mu m$ line using the radiation from a 29-GHz klystron to make up the difference, the first beat note observed was not this one. Instead, it was found experimentally to be between the $78-\mu m$ water vapor laser line, the third harmonic of the klystron frequency, and the simultaneous radiation of the 337- μ m lines of HCN. The only combination of harmonics which could produce such a coincidence, using frequencies calculated from wavelength measurements, is $6(\nu_{333}) - 2(\nu_{373}) + 3(29)$ GHz) = ν_{78} . The resulting frequency is 3.821775 ± 0.000003 THz. This number agrees remarkably well with that derived from the wavelength measurement quoted in Benedict et al. 9: 3.8218 ± 0.0003 THz. The frequency of the 373- μm line was measured by retuning the 29-GHz klystron so that $v_{337} - v_{373} = 3(29 \text{ GHz}) + 30 \text{ MHz}.$

The \pm 3 MHz uncertainty stems from inaccuracies in locating the maximum output of the laser line, which was determined by measuring the amplitude

of the beat note as the water vapor laser was scanned. The actual uncertainty in measuring the frequency was about 30 times less (about 100 kHz); it was due chiefly to the width of the beat note (about 50 kHz) and to some laser drift while the 29-GHz klystron was retuned to one-third of the difference between the 0.89- and 0.80-THz lines.

A representative signal from one of the better contacts producing the beat note from the $28-\mu m$ water vapor line is shown in Fig. 2. Although this signal appears to be only about 10 kHz wide, there is a frequency modulation of one of the lasers which synchronizes with the ac line, and the full width including this modulation is about 150 kHz. This beat note is derived from $12_{337} + 29$ GHz = $\nu_{28} - 35$ MHz with the result that $v_{28} = 10.718073 \pm 0.000002$ THz, in good agreement with wavelength measurements of Javan's group at MIT. 10 The same comments made about the accuracy and errors in the $78-\mu m$ frequency measurement are applicable in this case also, except that the center of the 10.7-THz line could be located somewhat more accurately.

For this measurement, the HCN laser was tuned to maximum output, and its frequency was found to be 0.8907606 ± 0.0000001 THz, in excellent agreement with the recent measurement of Hocker *et al.* 11

Obtaining the proper contact for the diode was an onerous task requiring an extremely delicate touch. The same contact worked equally well for the 78- and 28- μ m frequency measurement, and an earlier experiment showed another contact to work very well for the 12th harmonic of 74 GHz. Not only was the contact tenuous, but the adjustment of the microwave power level was extremely critical, and decreasing the 0.89-THz power by a factor of 2 completely submerged the beat note in the noise.

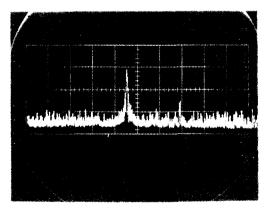


FIG. 2. Beat note between 28- μ m (10.7-THz) water vapor line and 12 times the 337- μ m (0.890-THz) HCN line plus 29 GHz from a klystron. The center frequency was at 38.3 MHz; the dispersion, 100 kHz/cm; and the sweep rate, 2 msec/cm. The signal from the 78 μ experiment was roughly equivalent to this one.

This particular point contact had been made many times; however, only when it had been left open for several minutes (possibly to oxidize?) were we able, after remaking the contact, to generate these high harmonics. The contacts were so fickle that they never remained overnight and often were destroyed with an interruption of the 10.7-THz radiation.

Work will be continued to compare various types of point-contact diodes including metal-on-silicon. It may be noted that tungsten-on-silicon contact in the same geometry produced up to 800 mV of detected 0.89-THz signal, measured directly on an oscilloscope, while the high-harmonic tungsten-on-nickel junction produced about 35 mV of opposite polarity.

In addition to providing a major step in extending the science of frequency measurement toward the visible region of the electromagnetic spectrum, the 78- μ m measurement suggests the possibility of combining the harmonics of several different laser lines and klystrons in order to reach any desired frequency to which the catwhisker will respond. Daneu et al. have shown that the metal-onmetal diodes respond to frequencies at least as high as that of the CO₂ laser by beating the pulsed $28-\mu$ m water vapor line with the pulsed $9.3-\mu$ line of CO₂. Attempts can now be made to measure one of the cw CO₂ lines by generating the third harmonic of the newly measured 10.7-THz line.

The authors would like to thank Professor A. Javan and Dr. L. Hocker for their stimulating discussions and for providing an accurate value of the wavelength of the 28-µm line. They would also like to thank G. Wichman and K. Gebert of the instrument shop for their excellent contributions to the project, Dr. J. Baird for suggesting various techniques of coupling the radiation to the catwhisker, and R. M. Charlton, M. Zanboorie, and Dr. F. Phelps for their assistance during several stages of the experiment.

 $^{^{1}}$ R. Barger and J. Hall, Phys. Rev. Letters $\underline{22}$, 4

²G. R. Hanes and C. E. Dahlstrom, Appl. Phys. Letters 11, 362 (1969).

³L. Frenkel, T. Sullivan, M. A. Pollack, and T. J. Bridges, Appl. Phys. Letters <u>11</u>, 344 (1967)

⁴L. O. Hocker, J. G. Small, and A. Javan, Phys. Letters 29A, 321 (1969).

⁵V. Daneu, D. Sokoloff, A. Sanchez, and A. Javan, Appl. Phys. Letters <u>15</u>, 398 (1969).

⁶L. O. Hocker and A. Javan, Phys. Letters <u>26A</u>, 255 (1968).

⁷In preparation.

⁸In preparation.